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[17]This $1^1S_0 \rightarrow 3^1P_1$ transition might not be observable if its width were significantly greater than 20 Å.

EXCITON-PHONON INTERACTION IN ALKALI HALIDES

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High-resolution optical studies of alkali halides at liquid-helium temperature have revealed complex structures associated with the exciton peaks. A sideband picture which involves the emission either of localized vibrations or of crystal phonons, depending upon the exciton radius, is proposed.

The fine structure of the exciton lines, which has been detected in some alkali halides,[1,2] casts new light on the role of the exciton-phonon interaction in these crystals. Until recently the exciton-phonon interaction in the alkali halides was assumed to cause only a broadening of the exciton lines.[3] In fact, according to the large values of the linewidths as obtained from thin-film absorption data, phonon effects could not be observed directly. However, the linewidths as obtained from Kramers-Kronig analysis of reflectivity data on single crystals are much narrower than those measured on thin films, and as a primary consequence fine structured excitons have been observed in several alkali halides.[2,4]

One can observe the structures which accompany the excitons in some alkali halides in Figs. 1-3, where we report the reflectivity spectra of NaBr, KI, and LiI cleaved in situ at about 6°K. The $n=1$ exciton is clearly split in NaBr, Fig. 1, while it appeared only as a shoulder at 55°K[2]; the magnitude of the splitting is 39 ± 3 meV. In KI, Fig. 2, the $n=1$ exciton shows a shoulder on the high-energy side, separated by 35 ± 5 meV, while only a slight asymmetry was visible at 55°K.[2] It is to be noted that in both cases the energy splittings of the $n=1$ exciton are well above the maximum phonon energy of the perfect crystal, which is 28 meV in NaBr and 18 meV in KI.[5] More striking is the shape of the $n=2$ exciton in KI, which shows a well-defined structure, composed of four or five peaks, with a separation of 16.5 ± 1 meV. A similar situation has been observed at 6°K also in RbI, with a splitting of 12 meV.[4] Several peaks are found also in the $n=1$ exciton of LiI as shown in Fig. 3. Their average separation is 39 ± 3 meV while the maximum phonon frequency of the crystal corresponds to 45 meV.[6] Multiple structure is observed also in NaI at liquid-helium temperature.[4]

According to the present data, it is found that the exciton-phonon interaction in the alkali halides appears more explicitly than expected and therefore the problem deserves further attention. There are essentially two mechanisms which describe this interaction: (1) Jahn-Teller instability when the exciton transitions take place between degenerate states; (2) phonon emission (or absorption) as found, e.g., for rare-earth ions.[7] The former model has been tentatively invoked in Ref. 1 in order to justify the doublet found in

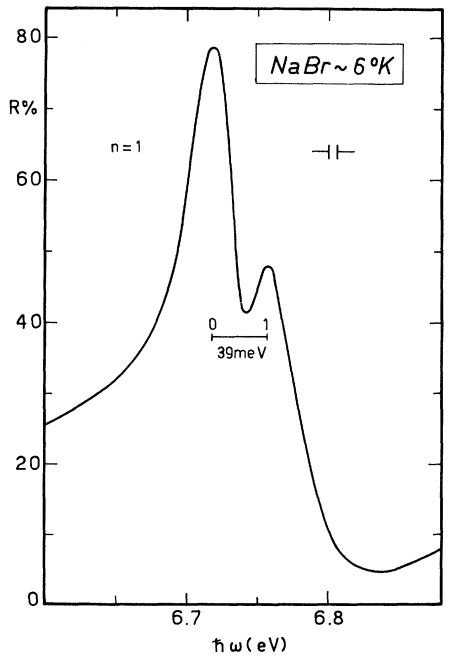

FIG. 1. Reflectivity spectrum of the lowest exciton peak ($n=1$) of NaBr (the separation between the 0-phonon and 1-phonon lines is indicated).

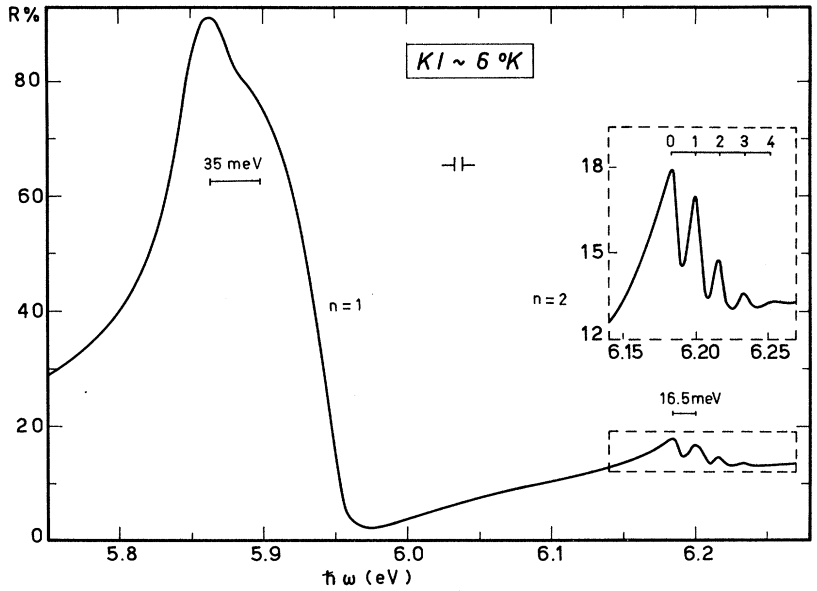

FIG. 2. Reflectivity spectrum of the $n=1$ and $n=2$ exciton peaks of KI. The $n=2$ exciton is also indicated in an expanded scale in the inset.

the lowest exciton of LiBr at 55°K. According to Ref. 1 the exciton degeneracy (hole at Γ_8^-), could be locally removed by vibrational modes of Γ_3 and Γ_5 symmetry, in a close analogy with the case of the F center in Cs halides.[8] The phonon sideband picture, on the other hand, has been introduced to explain also several fine-structured excitons in different crystals.[9] In the case of alkali halides its application may not appear so direct, since the energy separation, as discussed in Ref. 2, in some cases exceeds the maximum phonon energy of the perfect crystal. However, the multiplets reported here can be justified more directly in this scheme than in the Jahn-Teller picture which rather involves split lines.

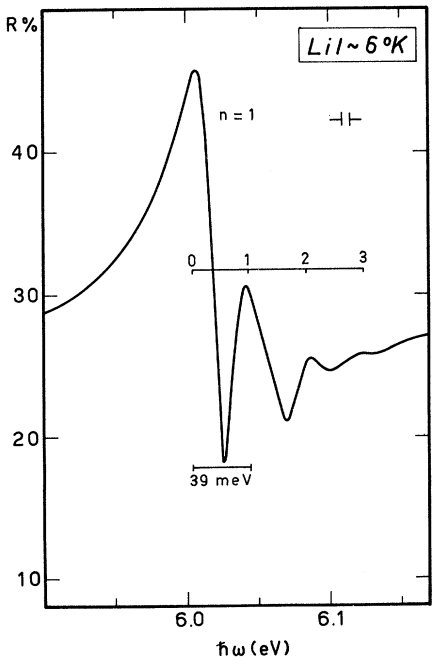

FIG. 3. Reflectivity spectrum of the lowest exciton peak ($n=1$) of LiI.

In the latter framework particularly instructive is the spectrum of KI since two excitons each accompanied by fine structure are observed at the same time. In the $n=1$ exciton the structure corresponds to phonons whose energy is clearly greater than that of the perfect-lattice phonons, whereas the $n=2$ exciton is followed by a few peaks whose average spacing is close to the LO-phonon energy. We may assume that small excitons (KI, $n=1$; NaBr, $n=1$) couple with localized phonons of frequency larger than that of the perfect lattice whereas larger excitons[10] (LiI, $n=1$; KI, $n=2$) are accompanied by crystal phonons.

The presence of high-energy vibrations for the case of small excitons indicates that the exciton itself perturbs the lattice strongly so that besides terms linear in the electron-phonon interaction, also quadratic terms occur and localized phonons of larger energy are created. The more extended excitons which cause locally even a

larger perturbation on the lattice dynamics are accompanied, instead, by perfect-crystal phonons since their spatial extension is such as to cover a larger region which is not appreciably perturbed by the exciton. The two cases examined here may in a way be analogous to the interconfigurational and intraconfigurational transitions of Sm^{++} ions in alkali halides.[11] We note also that the Huang-Rhys factor, i.e., the average number of phonons which accompany the electronic transitions, a measure of the linear coupling between electron and phonons, is larger for $n=2$ excitons in agreement with a similar observation in CuCl.[12,13] In fact the estimated values of the Huang-Rhys factor, i.e., the ratio of the first to the zeroth line, are approximately 0.1 for NaBr, 0.2 for LiI,[2] and 1 for KI, $n=2$.

A comparison with the projected densities of states relative to the lattice vibrations of KI and LiI[6] shows that the phonons emitted during the exciton (LiI, $n=1$ and KI, $n=2$) creation are of symmetry Γ_1. It is found that the peaks of the Γ_1 distribution, relative to the negative site, occur at ~16 meV for KI and ~36 meV for LiI in very close agreement with the values of the splitting reported above.

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DYNAMIC MEASUREMENT OF ELECTRON ENERGY RELAXATION IN InSb†

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We have measured the time dependence of the hot-electron current following a step variation in the applied electric field in n-InSb at 4.2°K. We obtain the average electron energy as a function of electric field, as well as the energy dependence of both the mobility and the energy relaxation rate.

Transport phenomena in strong electric fields are dependent on the details of the electron-phonon coupling. In this Letter, we report dynamic measurements of the electron-phonon relaxation in n-type InSb at helium temperatures. Our method[1] allows the direct determination of the electric field dependence of the mean electron energy ϵ, and consequently the energy dependence of both the mobility μ and the electron-phonon interaction.

One of the first studies of low-temperature transport phenomena in InSb was that of Sladek,[2] who estimated the electron-phonon coupling constants from the field dependence of the electron mobility. Preliminary dynamic measurements were carried out by several authors,[3,4] while more detailed results were reported by Kinch[5] and Lifshits, Oleinikov, and Shulman.[6] These measurements present conflicting views of the dominant energy exchange mechanism and are subject to certain experimental and theoretical difficulties which we shall discuss further on.

In the present work, we study the electron dynamics in the transition between one steady state and another. More specifically, we measure the time dependence of the current density j following a small step variation ΔE in the electric field E (see Fig. 1). The average energy satisfies the usual energy-balance equation

$$n\,d\epsilon/dt = \vec{E}\cdot\vec{j} - nP, \qquad (1)$$

where n is the electron density and P is the pow-